

Advances in Plant Mediated Iron Oxide Nanoparticles for Dye Colorant Degradation—A Review

Louisah Mmabaki Mahlaule-Glory and Nomso Charmaine Hintsho-Mbita * 

Department of Chemistry, University of Limpopo, Private Bag X 1106, Sovenga, Polokwane 0727, South Africa

* Correspondence: nomso.hintsho-mbita@ul.ac.za; Tel.: +27-(0)-15-268-2205

Abstract

Water polluted by dye colorants has been on the rise in the last decade. This is due to the over reliance on the textile industry, and it is holding a high economic value in most countries. This industry is the highest consumer of fresh water whilst also discharging several natural and synthetic pollutants to the environment. Several methods have been used for the removal of these pollutants and one of the most efficient technologies to be developed includes the photocatalysis method, via advanced oxidation processes. This review highlights the developments of green iron oxide nanoparticles as photocatalysts in the last decade. It was noted that tuning and controlling the phytochemical concentration and synthesis conditions, can assist with forming uniform and non-agglomerated materials, as this has limited the vast usage of these materials in major applications. Also, upon controlling the synthesis conditions, improved surface area and charge separation efficiency was noted. Their limitations and need for modification through forming composites are highlighted. Moreover, future perspectives are given on the use of green IONPs as photocatalysts.

Keywords: green iron oxide nanoparticles; dye colorant degradation; water treatment

1. Introduction

Color is one of the significant drivers of the fashion industry, in particular when deciding on the garment to purchase [1]. This high demand has increased the strain on the textile industry, which uses dyes for 95% of their garments. At present, close to a million tons of dyes are produced on a yearly basis, with 12% of them not being disposed properly. The textile dye market size was reported to be at 12.11 billion tons in 2023, which is estimated to grow by a further billion in a decade. Dyes, which are also known as colorants, are color imparting compounds [2,3]. These compounds can be natural or synthetic, depending on the source (Figure 1). They can be classified based on their chromophores (ability to impart color), ionic/non-ionic nature, and solubility [4]. For example, dyes, consisting of functional groups such as aniline, sulphonic acid, and hydroxides are known as auxochromes. Disperse dyes have been shown to be non-ionic and highly insoluble, whereas direct dyes exhibit excellent water solubility [5,6]. Of the dyes in use, reactive dyes, an example of a synthetic dye, is one of the most popular due to the varied colors it possesses and its impeccable wash fastness ability [7,8]. The second class that also exhibits the fastness ability is the vat type, which is known to be insoluble in water; however, these dyes could be transformed into a soluble solution when they are reduced by sodium dithionite, which act as a reducing agent in an alkaline solution to form vat acids/dyes (Indigo, Vat Blue 1, Vat orange, and many more) [9–11]. They are also



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observed by the presence of at least one keto group ($>C=O$), which allows them to show good fastness quality that exhibits high affinity for cellulose fibers. Thus, making them useful for coloring cotton materials/fibers such as denim, etc. [12,13].

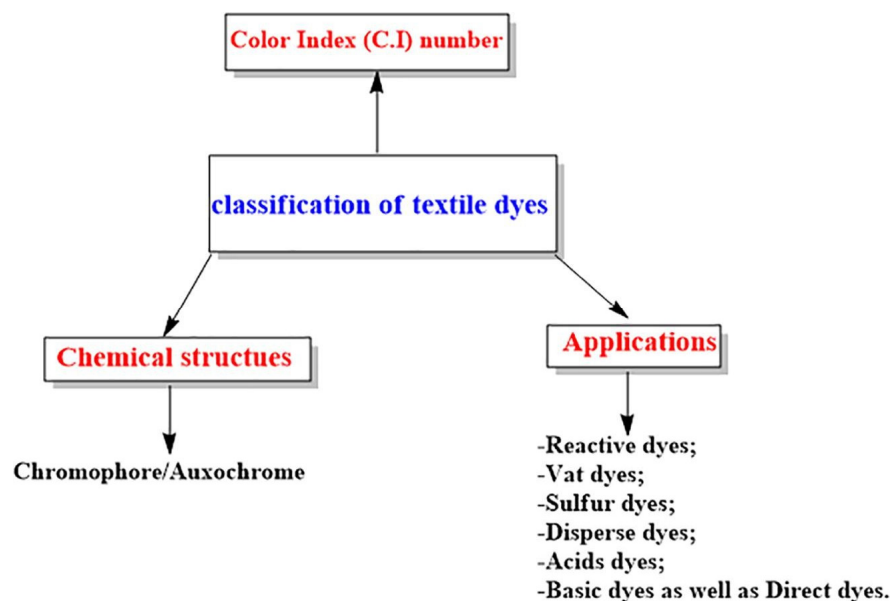


Figure 1. Classification of natural and synthetic dyes (OA) Revised and Reprinted from [14].

Though these dyes are of high usage, their impact on humans and the environment has been detrimental [6,15]. On a yearly basis, 14,000 humans die due to water polluted by dyes. The biggest damage is through leached untreated effluents being driven through the water stream [16–18]. A majority of these dyes are soluble in water, resulting in difficulty in removing them [7,19]. The color (Figure 2) linked to these dyes can cause lack of penetration in water, affecting the photosynthetic process. Also, in humans, they can be cancer causing agents, since they are mutagenic and carcinogenic, thus rigorous treatment strategies are required [8,20].

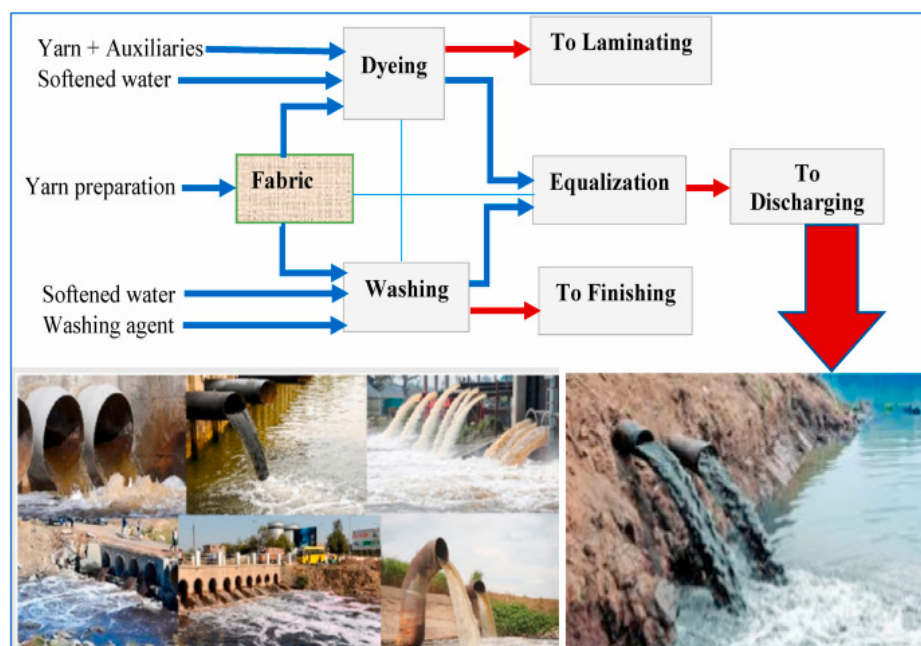


Figure 2. Textile industrial wastewater spreading on land and in an aquatic (OA) Reprinted from [20].

Several methods including reverse osmosis, solvent extraction, electrodegradation, adsorption, and membrane nanofiltration have been used for the removal of dye colorants in water [21,22]. For example, adsorption uses the adhesion of pollutants onto the surface of solid materials through physisorption or chemisorption [23,24]. This method is straightforward and has been used for the removal of different pollutants; however, it can be costly, requiring frequent regeneration of adsorbents [11,25]. In a study by Wong et al., using coffee waste modified with polyethylemine, they managed to adsorb ionic dyes reactive black 5 and Congo Red [6]. The adsorption took place via a mono layer adsorption and the chemisorption process [26–28]. A high adsorption capacity towards Reactive Black 5 (77.52 mg/g), compared to Congo Red (CR) (34.36 mg/g) was noted. This was caused by the high number of functional groups noted in Reactive black versus CR dye [6].

Biodegradation is another method on the forefront; it mainly uses bacteria and fungi to break down organic pollutants in a safe and efficient manner [29,30]. These microorganisms are introduced into wastewater in which they enzymatically degrade pollutants, using them as energy or nutrient sources [31–33]. However, the process can be slow, needing controlled environmental conditions and may not be effective for all types of pollutants [34–36]. In a study by Tian et al., [37] a fungal strain A-3 and bacterial strain L-6 were used for the degradation of crystal violet dye. In this study, a co-cultivation of the two strains was also investigated. From their analysis, the co-cultivated strain had a higher degradation efficiency compared to the mono-strains of fungi or bacteria. GC-MS indicated the presence of simpler, and safer organic compounds such as benzyl-alcohol, benzophenone, and benzaldelyde. This corroboration was confirmed via phytotoxicity tests showing less toxic products compared to the parent materials. Lastly, membrane filtration techniques have also been used to separate contaminants based on size exclusion or charge interactions [12]. The process includes choosing an appropriate membrane type based on contaminant size and properties (Figure 3). Wastewater is passed through the membrane, retaining contaminants while allowing clean water to pass [13,14]. This technique has high separation efficiency, capable of removing particles, microorganisms, and dissolved substances. Nonetheless, membrane fouling is a major limitation, in which the accumulation of contaminants on the membrane surface reduces efficiency [35]. Abid et al. [12] used RO and NFs membrane systems as an alternative method to remove acid red, reactive black, and reactive blue dyes from the Iraq mills. Variables such as temperature, pH, permeate flux and dissolved salts were investigated. At optimum conditions of 65 mg/L, 39 °C and a removal above 97% for all the dye colorants was noted. The NF method was more efficient. These methods have shown that based on the pollutant type and conditions presented, different methods can be employed. Researchers have recently geared towards the use of Advanced Oxidation Processes in the form of photocatalysis [38,39]. The aim of this method is focused on degrading pollutants into environmentally safe materials rather than removal [40].

Photocatalysis is a light mediated process whereby semiconductor materials such as Zinc Oxide (ZnO), Titanium Dioxide (TiO₂), Nickel Oxide (NiO) absorb photons and generate electron–hole pairs that can form redox processes [16,19]. In this process, light is used as a source of energy in the degradation of organic colorants into less harmful substances like water and carbon dioxide [20]. This process has been vastly used in particular for wastewater treatment.

In recent years, iron oxide nanoparticles (IONPs) have emerged as candidate materials in photocatalysis due to their high superparamagnetic surface reactivity and catalytic efficiency [21,22]. Green IONPs are not only safe, due to their low bandgap they also have access to the visible region. Moreover, compared to other traditional metal oxides, because

of their magnetic properties, they can be easily recovered from the wastewater system after treatment.

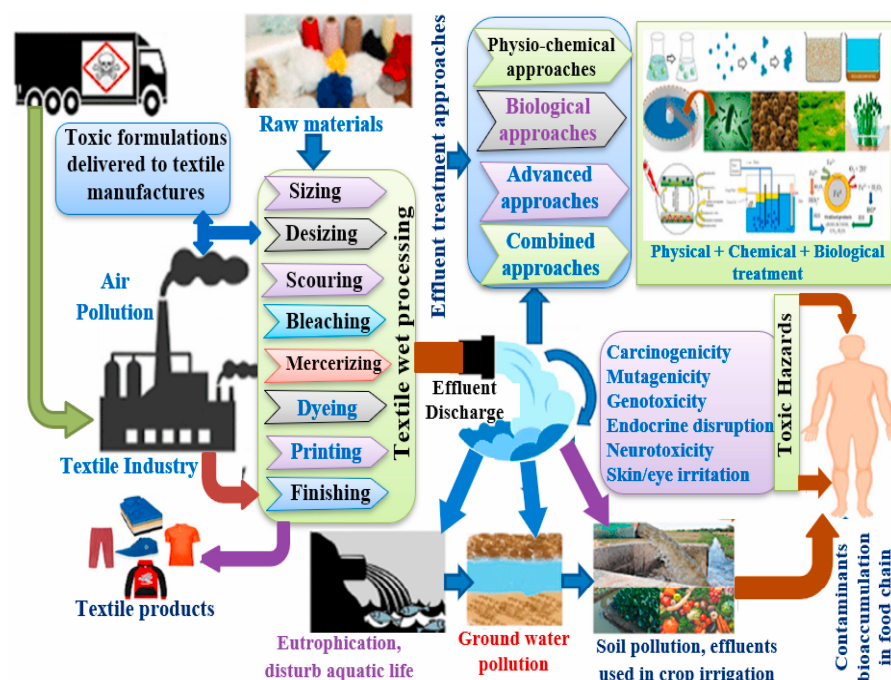


Figure 3. Impact of discharged dyes in water and the environment (OA) Reprinted from [20].

This review aims to highlight the current research on green iron oxide materials for the degradation of dye colorants in water. The focus is on green materials from the last 10 years, 2015–2025. The limitations of green IONPs photocatalysts will be highlighted, as will the strategies that have been gathered to overcome these. We will also look at the factors limiting these materials to be upscaled and used in high industrial corporations.

The novelty in the review lies in the fact that tuning and controlling the phytochemical concentration and synthesis conditions, the authors are able to assist with forming tailored, uniform, and non-agglomerated materials. Also, upon controlling the synthesis conditions, improved surface area and charge separation efficiency are noted. This is particularly the case when these materials are coupled with other materials allowing access to the visible region and enhanced degradation.

2. Iron Oxide NPs

Iron oxide nanoparticles (IONPs) are transition metal oxides that can come in different polymorphs of α -Fe₂O₃, Fe₂O₃, Fe₃O₄, and FeO [23]. The most common polymorphs include hematite (α -Fe₂O₃) and magnetite (Fe₃O₄).

Hematite (α -Fe₂O₃) is one of the most widely utilized iron oxide phases in photocatalysis due to its narrow bandgap that ranges between ~1.9 to 2.2 eV, which enables efficient absorption of visible light [7]. It is abundant, chemically stable, and environmentally benign, making it an attractive catalyst for sustainable applications. When their structure has been modified, optical properties and surface areas can be enhanced, thus assisting with the availability of the active sites and access to a wide range spectrum, which in turn can improve the photocatalytic nature of the materials [23]. In a study by Bibi et al., hematite (α -Fe₂O₃) nanoparticles, derived from *pomegranate* seed extract, achieved a 95% degradation against Reactive Blue dye in less than an hour under solar irradiation [24]. This performance suggested that the dye acted as a chromophore, absorbing visible light and injecting energetic electrons into the conduction band of hematite, thereby initiating photocatalysis.

These photo-excited electrons were subsequently captured by the catalyst's pores and surface-active sites, promoting efficient oxidation of the dye [24]. Overall, the interaction between the chromophoric dye and hematite nanoparticles underscored the role of dye-sensitization mechanisms in enhancing visible-light-driven photocatalytic degradation.

Magnetite (Fe_3O_4) is another polymorph of iron that has been widely used in catalytic applications. This is a ferrimagnetic iron oxide with an inverse spinel/cubic structure as noted in Figure 4b. It is known for its high electrical conductivity and facile magnetic recovery using external magnets [25]. This is a highly desirable feature for post-treatment catalyst separation [26,33]. Despite exhibiting a moderate optical bandgap of between 1.8 and 2.2 eV, pure magnetite demonstrated limited intrinsic photocatalytic efficiency owing to its restricted visible-light absorption range [27]. Nevertheless, the presence of mixed-valence iron states ($\text{Fe}^{2+}/\text{Fe}^{3+}$) enhanced its electrical conductivity and facilitated the charge transport, making it well-suited for synergistic redox systems and coupled photocatalytic reactions [26].

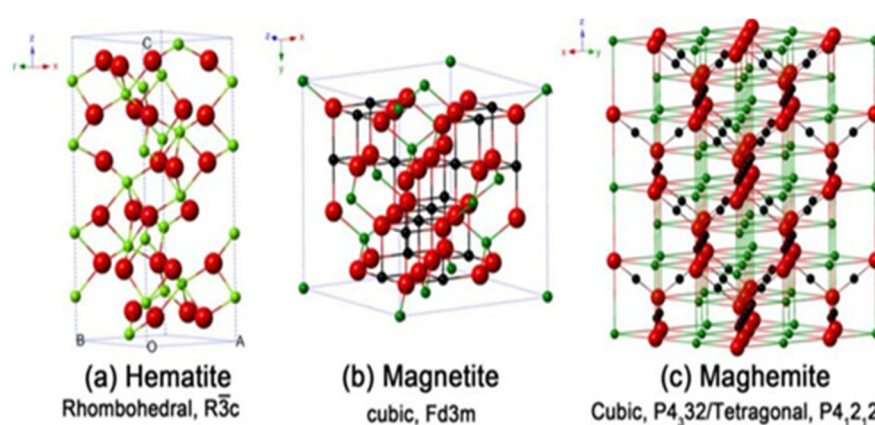


Figure 4. The three crystal structure of IONPS (a) Hematite, (b) Magnetite and (c) Maghemite (OA). Reprinted from [19].

Maghemite ($\gamma\text{-Fe}_2\text{O}_3$) is another form of a ferrimagnetic iron oxide with a spinel structure, and an optical bandgap of 2 eV [28]. It is often synthesized using the co-precipitation method followed by mild oxidation. This material offers chemical and magnetic properties that are favorable for catalyst recovery [29,30]. Sheikholeslami et al. [31] discovered that upon testing their catalytic efficiency against BTEX compounds under UV and visible light, at optimal conditions of pH 3, a concentration of 170 mg/L, the UV-assisted degradation produced a 97% degradation in 90 min compared to the visible light assisted degradation of 95% over a 5 day period. From these materials, it has shown that (IONPs) Ion oxide NPs have strong catalytic abilities. However, the usage of toxic reducing agents in particular for biological and environmental applications might hinder their overall usage, especially when upscaling; thus, researchers have geared towards the usage of green materials using plant extracts.

3. Green Synthesis of Iron Oxide NPs

The synthesis of iron oxide nanoparticles, in particular through the use of plant extracts has been the most widely used compared to the usage of micro-organisms like fungi and bacteria [32]. Plant extract-mediated synthesis uses water soluble phytochemicals as reducing and capping agents. These include proteins, polyphenols, alkaloids, sucrose, phenolic acids, and co-enzymes [33,34]. By reducing Fe^{3+} from salt precursors like $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (iron(III) nitrate nonahydrate) and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (Iron(III) chloride hexahydrate) to Fe^0 from these phytochemicals when their functional groups interact with these salts. Thus, form-

ing hematite ($\alpha\text{-Fe}_2\text{O}_3$) through controlled synthetic parameters like (pH, calcination and heating temperatures, type of plant, and many more). When these conditions are met, the morphology, surface chemistry, crystallinity, and particle size of the materials are therefore controlled. This was observed when a study by Bouafia and Laouini [1] where the effect of ferric chloride concentration using the extract of *Metha pulegium* L. was undertaken. Two types of cubical shaped materials (Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$) were formed. These were confirmed with the Fe-O stretch at 510 cm^{-1} and an excitation between 275 and 301 nm. Ahmed et al. [3] in the presence of *Actinidia chinensis* extract also formed cubic and rod-shaped IONPs with a particle size range of 92–107 nm. The extract used as a reducing agent for the synthesis, contained polyphenols, flavonoids, and phenolic acids, which had large aromatic hydroxyl groups and singlet oxygen quenchers [Figure 5]. In another study, using *Vernonia amygdalina* leaf extract, the Fe-O bond at 554 cm^{-1} corroborated the formation of these FeNPs. Their materials were non-uniform, amorphous, and highly irregular. Khadka et al. [13], using *Senegalia catechu* also formed the pure, uneven, and spherical $\alpha\text{-Fe}_2\text{O}_3$. XRD confirmed the phases formed and the crystallite size to be 34 nm. Furthermore, the UV peak excited at 580 nm, was reflective of the transition of Fe^{3+} ions and these are due to 3D orbitals. Goudjil et al. [16], studying the effect FeCl_2 concentration, using *Mentha pulegium* extract, managed to form hexagonal $\alpha\text{-Fe}_2\text{O}_3$ with a direct bandgap size ranging from 3.2 to 3.9 eV and particle size of 16–35 nm. The authors stated that the strong absorption peak noted at 290 nm was due to the collective oscillation of the electrons from the conduction band. Furthermore, they stated, the Fe^{2+} ions were converted into Fe^0 during the reduction stage [36].

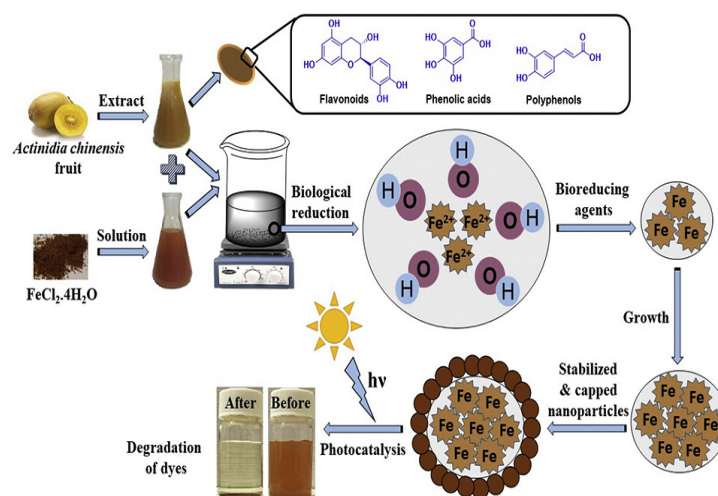


Figure 5. *Actinidia chinensis* derived IONPs and their synthesis path. Reprinted with permission from [3].

Yassin et al. [33] in the presence of *Cammellia sinesis* synthesized amorphous Fe_2O_3 NPs with a particle size of 49 nm and a surface charge of $-48.5\text{ m}^2/\text{g}$. The UV peak at 241 nm confirmed the formation of the material. Dash et al. [34] also synthesized mesoporous magnetite NPs using *Peltophorum pterocarpum*. Fe-O bands at 557 cm^{-1} , confirmed the formation of the spherically shaped material with a crystallite size of 23 nm and surface area of $47\text{ m}^2/\text{g}$. Ekwumemgbo et al. [35] further formed spherical morphology Fe_3O_4 *Proponis africana* mediated NPs with a similar surface area of $47\text{ m}^2/\text{g}$. A UV peak at 400 and 633 nm also corroborated the formation of these particles. In the presence of *Briddellia retusa* leaf extract, hematite spherically shaped magnetic Fe_2O_3 NPs were formed. A high surface area of $75\text{ m}^2/\text{g}$ compared to the previously synthesized green materials was formed. From this study, the authors postulated that at first the Fe^{2+} ions were reduced

by the phytochemicals (OH) from the plant to zero valent ions. Thereafter, oxidation to form Fe^{3+} took place. This was followed by aggregation and nucleation leading to the production of Fe_2O_3 . Vinayagam et al. [38] also formed a high surface area ($191 \text{ m}^2/\text{g}$) super paramagnetic rod like Fe_2O_3 material (Figure 6). The small particle size of 11 nm may have assisted in the high surface area. Furthermore, the iron material demonstrated high thermal stability with a magnetization of 34.43 emu/g . Their formation was further confirmed through XPS with an oxidation state of Fe and O.

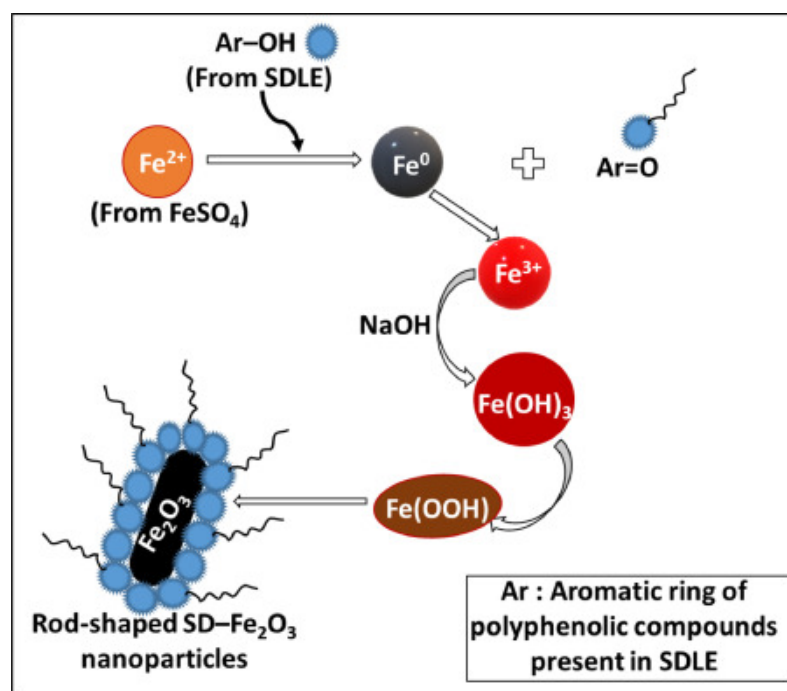


Figure 6. Mechanism of formation of rod-like Fe_2O_3 NPs via SDLE plant extract. Reprinted with permission from [38].

Magnetite is also synthesized by the same principle of reducing the salts from their functional groups through various phytochemicals obtained from different plants. In this type of synthesis, two sources of Fe (Ion) are required (Fe^{2+} and Fe^{3+}) as 1:2 ratio from a variation in salts precursors, using coprecipitation method for enhance magnetism. This is observed by a study performed by Mahlaule-Glory [39], where they used ferrous salt of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (Iron(II) sulfate heptahydrate) as the Fe^{2+} and ferrite salt $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (Iron(III) chloride hexahydrate) as the Fe^{3+} source with a ratio of 1:2 which also formed Rod like mesoporous magnetic magnetite NPs. However, their surface area was far smaller ($12.83 \text{ m}^2/\text{g}$). In their synthesis, they noted that a two-step reaction mechanism took place where phytochemicals consisting of hydroxides (OH) and aldehydes (R-CH=O) groups assisted in the reduction of Fe^{3+} to Fe_3O_4 NPs. Further, they noted, the hydrolysis process took place, proceeded by releasing H^+ ions. In a recent study, by Alprol et al. [40], formed irregularly shaped materials were aggregated and had clusters. Upon forming the magnetic material (Fe-O stretch bond), a negative surface charge (-6.32 mV) was noted, and the authors further elaborated that the chlorophyll found in the plant extract served as an electron donor for the reduction of Fe^{3+} to Fe^{2+} . Mukherjee et al. [41] also formed non-uniform aggregated spherical morphology materials with a particle size of 20–50 nm and a surface area of $23 \text{ m}^2/\text{g}$ (Figure 7a,b). They noted that the oxygen bound functional groups increased the nucleation sites, promoting the formation of the spherical NPs. Also, through XRD (Figure 7c), the rhombohedral structure coupled with the various phases formed confirmed the formation of this material. This was further corroborated by FTIR, where the

Fe-O stretch was identified in the fingerprint region of 470 cm^{-1} . Moreover, from UV-Vis, an excitation at 220 nm was noted, which resulted from the excitation of surface plasmon, where there was charge transfer from $\text{O}^{2-} 2p$ to $\text{Fe}^{3+} 3d$. Mbachu et al. [42], using *Cola nitida*, also confirmed the formation of magnetic IONPs via the Taguchi optimized method. The Fe-O stretch noted at 475 cm^{-1} and 620 cm^{-1} , was indicative from other studies [39,40]. A high surface area ($125\text{ m}^2/\text{g}$) with type IV isotherm was an indication that the material will have increased active sites.

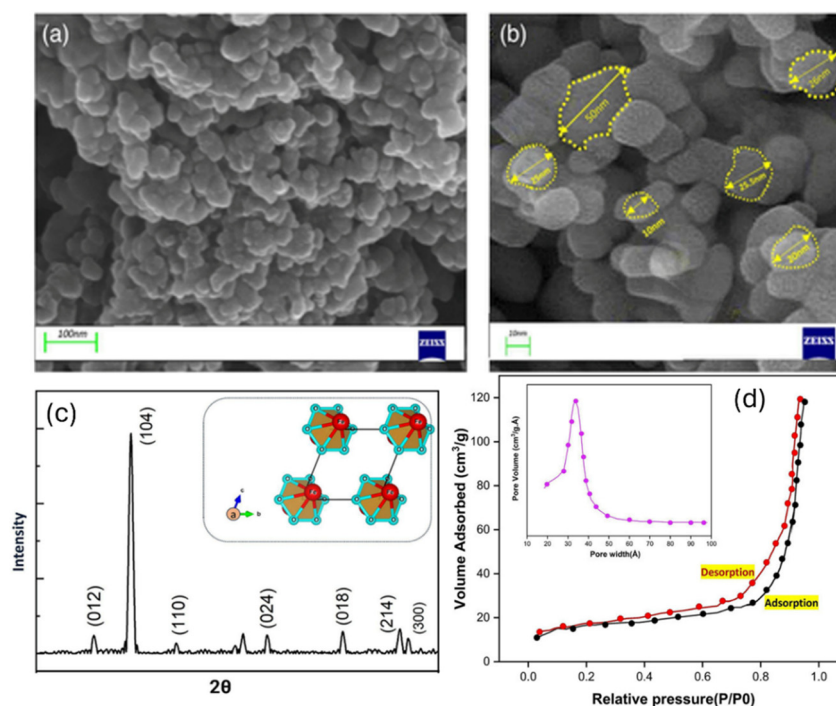


Figure 7. (a,b) SEM, (c) XRD and (d) BET of green Fe_2O_3 NPs. Reprinted with permission from [41].

Maghemite ($\gamma\text{-Fe}_2\text{O}_3$) has likewise been synthesized through oxidation of Fe^{2+} ions from salts to Fe^{3+} resulting in ($\gamma\text{-Fe}_2\text{O}_3$) with the interaction of phytochemical-mediated functional groups from plants, during heating or drying in the presence of air [43,44]. A study reported by Siddiqua et al. oxidized $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ to form $\gamma\text{-Fe}_2\text{O}_3$ from which a nanocomposite was synthesized and resulted in spherical morphology with 14 nm particle size [45].

Researchers have recently opted to use the green-synthesized IONPs due to the fact that they are sustainable, eco-friendly, and have triple R's as compared to the IONPs synthesized by chemical methods. The costs, secondary pollution, use of toxic reagents, higher maintenance requirements, expensive and higher synthetic conditions, limit the usage of these traditionally synthesized materials. Furthermore, the use of phytochemicals extracted from plants also act as capping/reducing and stabilizing agents. These phytochemicals possess functional groups that can modify the surface of IONPs and enhance their stability, help to prevent agglomeration, and improve catalytic interactions between the catalyst and the dye during photocatalysis. The modified surface will therefore influence the photocatalytic properties by facilitating better electron transfer and by increasing the active sites on the surface of IONPs. Whereas with chemically synthesized IONPs that often involve the use of toxic reagents like (NaBH_4 , N_2H_4 , H_2O , NH_3 , NH_4OH , NaOH , EDA , etc.), which are hazardous, use high energy inputs and toxic solvents, thus, resulting in non-ecological friendly material. Furthermore, their produced IONPS comes with limited surface functional area, requiring additional capping agents to achieve stability. Thus, the particles synthesized from these methods are then prone to exhibit higher agglomer-

ation tendencies, with lower biocompatibility, and reduced surface activity compared to green-synthesized IONPs.

In a study performed by Meyyappan et al. [46], a comparative study was conducted between green and chemically synthesized IONPs. During the synthesis phase, the green synthesized IONPs were formed using a shrub (*Desmodium gangeticum*) DG at lower temperature, whereas with the chemical synthesis method, ammonia was used to synthesize IONPs. These two materials were further characterized and were confirmed to both yield similar results in terms of their characterization techniques. However, in terms of toxicity, they further tested these two materials against normal cells, and it was discovered that the chemically synthesized IONPs were more toxic and would have the potential to damage human organs.

The literature has reported that the degrading efficiency of IONPs is influenced by additional variables such as surface chemistry, porosity, particle size and shape, crystallinity, band gap, light absorptivity, and charge separation. To evaluate the efficacy of the chemically or biologically synthesized IONPs, a variety of published studies were examined and compared based on these parameters. The first one was reported by Adekunle et al. [47] when reporting on a comparative study of microwave and chemically synthesized IONPs for the degradation removal of two organic dyes, namely murexide (98%) and EBT (96%), at each dye's optimal conditions [47]. According to their reports, the microwave synthesized IONPs gave good removal efficiency, regardless of the dosage loading. Then, they also performed similar photocatalytic degradation and discovered that lower removal efficiency was recovered when the chemically synthesized IONPs were used. The report suggested that the material did not improve efficiency; however, it indicated that at lower catalyst loadings of 5 mg (15.2%) and 15 mg (21.4%) for each dye, respectively. From this study, this degradation removal % was obtained at lower catalytic loading. Suggesting that the obtained surface area could have been lower, hence it affected the degradation of the chemically synthesized IONPs at higher catalytic loading of IONPs. This might be assumed that the aggregation would have occurred, and the active sites would have been blocked due to saturation at higher dosages [47]. Moreover, this suggests that the recombination rate was higher, hence fewer carriers were observed particularly for the chemically synthesized IONPs. According to their findings, the microwave showed better degradation removal than the chemically synthesized IONPs [47].

In another study, Qasim et al. [48] compared chemically and biosynthesized IONPs, and according to their report, it revealed that the green synthesis material had more degradation efficiency than the chemically synthesized material. The report did not disclose the actual results but rather reported on the enhanced effectiveness of the green synthesized IONPs compared to the chemically synthesized IONPs. Furthermore, the toxicity of the green synthesized IONPs was also examined in the same research study, and it was shown to be less toxic than the chemically synthesized IONPs [48]. This is compared to green synthesized IONPs reported by Khatun et al. [23], Jara et al. [5] and Chaundhari et al. [24] where their degradation efficiency was reported to be 100%, 95%, and 97%, respectively, against various dyes. This could have been due to plant extracts that could have generated higher surface area and reduced the recombination rate. Moreover, more electron-hole pairs with more active sites on the surface material would have been generated. Further, expecting enhanced degradation efficiency [38].

From Table 1, we have noted that the most stable form of IONPs, α -Fe₂O₃, was the most dominant polymorph formed [37]. These materials were noted to be spherical, rodlike, polydispersed, or agglomerated. In one study, they noted that the high formation of the spherical morphology was due to iron materials having a basal polar oxygen face and an overexposed polar M-metal. Also with green materials, the phytochemicals used for

reducing these metals tend to also play a capping and stabilizing role, thus, preventing aggregation and enhancing the formation of a spherical morphology [49,50].

Table 1. Plant-mediated synthesis of iron oxide nanoparticles and their characteristics.

Catalyst	Plant	Morphology	Particle Size (nm)	Surface Area (m ² /g)	Ref.
Fe ₃ O ₄ and α-Fe ₂ O ₃	<i>Mentha pulegium</i> L.	Cubical	22–34	-	[1]
Fe NPs	<i>Actinidia chinensis</i>	Cubic and Rod shaped	92–107	-	[3]
Fe NPs	<i>Vernonia amygdalina</i>	Polydispersed	2.31 μm	-	[5]
Fe NPs	<i>Senegalia catechu</i>	spherical	34.7	-	[13]
α-Fe ₂ O ₃	<i>Mentha pulegium</i>	Hexagonal shape	16–35	-	[16]
Fe ₃ O ₄	<i>Psidium guajava</i> L.	Cubic, partly spherical	12.64	-	[17]
FeNPs	<i>Mystrica. fragrans</i>	Polydispersed Quasi Spherical	20–45 nm	-	[22]
Fe ₃ O ₄	<i>Baccaurea ramiflora</i>	Agglomerated spherical	8.83 nm	-	[23]
α-Fe ₂ O ₃	<i>Pomegranate seeds</i>	Semi spherical and agglomerated	25–55 nm	-	[24]
Fe ₃ O ₄ and Fe ₂ O ₃	<i>Eregia densa</i>	Oval and cluster Oval/hexagonal	18.49	-	[27]
Fe ₂ O ₃	<i>Camellina sinesis</i>	Spherical	49	48.5	[33]
Mesoporous magnetite	<i>Peltophorum pterocarpum</i>	Spherical	23 (XRD)	47	[34]
Fe ₃ O ₄	<i>Prosopis africana</i>	Spherical	-	47	[35]
Fe ₂ O ₃	<i>Bridellia retusa</i>	spherical	38	75	[36]
α-Fe ₂ O ₃	<i>Spondias dulcis</i>	Rod like	11.38	190.84	[38]
Fe ₃ O ₄	<i>Monsonia burkeana</i>	Rod like	-	12.83	[39]
Fe ₂ O ₃	<i>Arthorospira platensis</i>	Irregular Clusters	50 μm	-	[40]
α-Fe ₂ O ₃	<i>Chrysanthemum indicum</i>	Aggregated spherical	20–50	23	[41]
Magnetic Fe ₃ O ₄	<i>Achyranthes. aspera</i>	Irregular spherical	5–60	-	[51]
α-Fe ₂ O ₃	<i>Cola nitida</i>	Spherical aggregates	-	125	[42]

In other instances, such as a high reaction temperature and when materials are over capped-high plant concentrations, aggregates might also form, where these phytochemicals chose to sit on top of the surface of the particles instead of blending with the salts [52]. These agglomerated NPs might also indicate strong molecular forces, in particular for H⁺ bonding between the OH groups and other phenolic compounds [53]. In terms of crystallite size and surface area, there was no direct correlation, as most of these materials were measured to be in the nanometer range. Also, few studies reported on the surface area, they mainly ranged between 30 and 80 m²/g but a study by Vinayagam et al. [38], which had the smallest particle size, produced the highest surface area. To further understand these materials and their efficiency, we will explore their ability as photocatalysts.

4. Green Iron Oxide Nanoparticles as Photocatalysts

The photocatalytic efficiency of green-synthesized IONPs has been widely studied, and the efficiencies have been shown to mostly depend on optimization of key operational

variables, including dye concentration, pH, catalyst dosage, and contact time [13,39]. In addition to these, the literature reports emphasize the influence of physicochemical properties such as light absorptivity, porosity, crystallinity, band gap energy, and surface chemistry on photocatalytic performance [13,27]. These factors collectively govern the generation and activity of reactive species during the photodegradation processes [54], which tailors these characteristics into an essential advancement that is efficient, stable, and uses sustainable photocatalysts aimed at environmental remediation applications [27,55].

During photocatalyst, when IONPs are used as photocatalysts to degrade various dyes, it is essential to understand the mechanistic interactions between each type of IONPs and the dye class involved, as displayed in Table 2. Organic dyes have unique chromophores, and these structural distinctions have a significant impact on their degradation processes. As a result, generally, dyes do not respond the same during photocatalysis. Consequently, dyes respond differently based on the nature of their chemical structures. For example, the azo dyes type possesses a chemical structure that contains a unique azo chromophore that contains the nitrogen triple bond ($-N=N-$), which is responsible for their bright color. Therefore, in photocatalysis, reactive radical species formed on the IONP surface attack the azo bond. These include the hydroxyl radicals ($\bullet OH$) and superoxide radicals (O_2^-) that attack and break the $-N=N-$ link. This is therefore visible within the conductive band when the oxidative reaction is triggered by electrons. Generating the photogenerated holes (h^+) within the valence band on the surface material when reducing water (H_2O) and producing $\bullet OH$ radicals. Consequently, the superoxide radicals form within the holes when the electrons degrade dissolved oxygen. These reactive species work together to disrupt the azo bond, resulting in dye degradation and generation of less harmful mineralization of CO_2 and H_2O . This is observed when a study performed by Truskewycz et al. [2], where a green tea was used to mediated IONPs and managed to degrade 90% of Azo dyes mixtures at a concentration of 0.53 ppm within 20 min. The efficiency was pH and temperature dependent, favoring lower pH levels at a temperature range from 70 to 90 °C. In this study, it appeared that the interaction between the azo functional groups of nitrogen bonds ($-N\equiv N-$) were attacked by the radical species into smaller and simpler ions by the degradation of the dye. Hence, a higher degradation removal percentage was obtained. The same principle is also observed in a study reported by Ahmed et al. [3] where 94% of Alizarin Yellow was degraded using sunlight over a 4 h period [3]. The OH radicals were noted to be the responsible species to attack and destroy the azo bond and degrade the dye, and also the first order kinetics were recorded. Furthermore, in addition to the effectiveness of the radical species, there is also a factor of a large surface area that can be seen in the synthesis of plant-based IONPs. That generates more binding sites and more electron-hole pairs that minimize the recombination rate and also increases the photocatalytic efficiency of the IONPs surface. Hematite and magnetite surface chemistry are known to be good absorbers of UV radiation, regardless of the source. Be it from the sunlight or the UV-lamps, they tend to absorb light and increase the degradation efficiency. These factors might have contributed to a study reported by Jara et al. [5] where they managed to obtain a degradation of 97 and 94% for CV and MB dyes, respectively, with a correlation of close to 1 (0.999 and 0.995) in both reactions. Upon testing the recyclability, the material could still be used for more than four cycles with an efficiency of above 90%.

Table 2. Green synthesized IONPS for the degradation of various colorants using photocatalytic applications.

Type of IONPs (Catalyst)	Dye Class & Chromophore	Dye Type	Light Source	Optical Properties	IONPs Conc	Dye Conc	Other Parameters	Contact Time	Degradation (%)	Reuse	Ref.
Fe	Basic Dye <i>Azo</i>	AYR	Sunlight	-	100 mg/L,	100 ppm	pH = 6	4 h	94	-	[3]
Fe	Reactive dye <i>Anthraquinone</i> Direct dye <i>Azo</i>	RBB RD80	UV 0.75% H ₂ O ₂	-	150 ppm 25 ppm	0.53 ppm	-	20 min	90	-	[2]
Fe	Acid dye <i>Azo</i>	MO	UV light, (Hg Lamp) 500 W	293 nm	30 mg	25 ppm	-	90 min	95	5 cycle	[21]
Fe	Basic Dye <i>Triarylmethane</i> Basic dye <i>Phenothiazine</i>	CV MB	H ₂ O ₂	-	0.01 → 0.125 mg	5–30 ppm	pH = 2–12	30 min	97 and 94	4 times	[5]
FeO	Basic dye <i>Xanthene</i>	RhB	UV	-	-	-	-	60 min	95	-	[55]
FeO	Basic Dye <i>Triarylmethane</i>	CV	Sunlight	-	10 ppm	-	-	150 min	80	-	[56]
FeO	Acid dye <i>Azine</i>	S	Sunlight	294 and 278 nm	0.5 mg	1 ppm	-	180 min	-	-	[48]
α-Fe ₂ O ₃	Basic Dye <i>Phenothiazine</i>	MB	Sunlight	2.2 eV 580 nm	5 mg	4 ppm	-	~120 min	~80	3 cycles	[13]
IONPs	Acid dye <i>Azo</i>	MO	UV light	-	10 mg	10 ppm	-	90 min	>70	-	[14]
α-Fe ₂ O ₃	Direct dye <i>Azo</i> Direct dye <i>Di-azo</i>	CR EB	UV light	—	2 mg	10 ppm,	pH = 8	—	-	-	[15]
α-Fe ₂ O ₃	Basic Dye <i>Phenothiazine</i>	MB	Photo reactor (5 Watt)	3.2 eV	10 mg	10 ppm,	pH = 6.5; 250 rpm	120 min	78	-	[16]
α-Fe ₂ O ₃	Basic Dye <i>Phenothiazine</i> Acid dye <i>Azo</i>	MB MO	Light + H ₂ O ₂	—	1 mg	25 ppm	pH = 6,	95 min 205 min	82 54	-	[17]

Table 2. Cont.

Type of IONPs (Catalyst)	Dye Class & Chromophore	Dye Type	Light Source	Optical Properties	IONPs Conc	Dye Conc	Other Parameters	Contact Time	Degradation (%)	Reuse	Ref.
α -Fe ₂ O ₃	Phthalein dye Triarylmethane Direct dye Di-azo	PR EB	35 W UV Lamp	220 nm 2.6 eV	1.5 g,	10 ppm	-	-	98 90	5 cycles	[41]
Fe ₂ O ₃	Basic dyes Phenothiazine Basic Dye Triarylmethane	MB CV	Light source 250 W	-	0.1 g	20 ppm,	450 rpm,	180 min	66 66	-	[18]
Fe ₂ O ₃	Direct dye Azo dye	RR	Sunlight	-	0.8 g/L	70 ppm	pH = 2	6 h	77	-	[32]
Fe ₂ O ₃	Basic dye Xanthene	Rhb	UV light	-	50 mg,	20 ppm	-	56 min	95	-	[24]
Fe ₂ O ₃	Basic Dye Phenothiazine Acid dye Azo	MB MO		2.84 eV	25 mg/L	100 ppm,	pH = 5, 50 °C	60 min	88 82	6 cycles	[42]
Fe ₂ O ₃	Basic Dye Phenothiazine	MB		2.84 eV	10 mg,	10 ppm,		210 min	93	-	[28]
Fe ₃ O ₄	Basic dye Phenothiazine	MB	Sunlight & Hg lamp (9 W)	-	12 mg	25 ppm	pH = 12	50 min	82 94	-	[57]
Fe ₃ O ₄	Basic Dye Phenothiazine	MB	UV)	−300 nm	40 mg	10 ppm	pH = 7,	20 min	100	-	[23]

MB = Methylene Blue, CV = Crystal violet, AYR = Alizarin yellow R, RR = Remazol yellow, S = Safranin, RB = Reactive blue, RhB—Rhodamine Blue, CR = Congo Red, MO—Methylene Orange, EB = Evans Blue, PR—Phenol Red, RBB = Remazol Brilliant Blue, DR80 = Direct Red 80.

Other researchers prefer to use the photocatalysis process to decompose dyes and H_2O_2 in the presence of iron (Fe^{2+} or Fe^{3+}) to generate effective radical species used to degrade dyes and this was also observed in a study reported by Khadka et al. [13]. They used sunlight and assisted by H_2O_2 and managed to degrade 80% of MB after 120 min at optimum conditions. However, in the absence of H_2O_2 in this study it was discovered that green synthesized IONPs as a catalyst managed to MB by 85% after 7 h. Therefore, according to their finding, it was reported that the addition of H_2O_2 shortened contact time from 7 h giving 85% to 2 h giving 80%. This was possible by the addition of H_2O_2 that generated $\text{OH}^\cdot$ radicals that attacked MB dye. The authors suggested that H_2O_2 had acted as an electron scavenger by improving the electron-hole separation, which minimizes the recombination rate and thus, encouraging the production of reactive oxygen and radical species. This synergistic effect tends to eventually increase the photocatalytic degradation efficiency [5,13]. After reusing the materials for four cycles, a sharp decline in the degradation was noted to 54% in the fourth cycle. This could be due to stability and prolonged service life. Adhikari et al. [17], with a small particle size of 12.6 nm managed to degrade 82% of MB and 54% of MO after 95 min. A red shift from 621 to 630 nm was recorded for MB. The authors stated that it could be caused by MB protonation, thus decolorization of the MB in a fenton-like reaction.

Radin et al., [21] with zero valent Fe^0 NPs through TGF extract, managed to degrade more than 95% of MO dye in less than 2 hrs. Reusability studies showed the material could be re-used up to five times, influenced by first order kinetics with a rate constant of $k_{\text{app}} 0.025 \text{ min}^{-1}$. Khatun et al., [23] managed a completed degradation/decolorization of MB in 20 min using spherically shaped Fe_3O_4 NPs with a particle size of 8 nm. The small particle size could have assisted in the efficient degradation as a rate constant of $0.067 \pm 0.010 \text{ min}^{-1}$ was noted. Matar et al. [28] used IONPs derived from *Brown Egyptian Propolis* (BEP) extract against MB. At an optimized concentration of 10 ppm, a 93% degradation efficiency in 210 min was noted. From the study it was concluded that as the dye concentration increased to moderate levels, a saturation effect began to emerge wherein the dye molecules increasingly occupied the photocatalyst surface, thereby reducing the number of available active sites for photocatalytic reactions.

In photocatalysis, pH plays an important role in the degradation of various dyes. It is governed by the interaction between the dye and the catalyst. Electrostatic ionization and many other influences, surface chemistry, dispersion stability, and the ionization of the material occur when the two interact. This is observed when the interaction occurs when the conditions are either acidic or alkaline, where dyes may undergo protonation or deprotonation on the photocatalyst surface and thus, influencing degradation efficiency.

Mukherjee et al. [41] used a 35 W UV-assisted magnetic $\alpha\text{-Fe}_2\text{O}_3$ nanoparticles from *Chrysanthemum indicum* flower extract to degrade Phenol Red (PR) and Eosin Yellow (EY). A high efficiency of 98% and 90% for PR and EY dye in 90 min was noted. The high efficiency could be due to the low bandgap of 2.6 eV. The authors stated that when the green $\alpha\text{-Fe}_2\text{O}_3$ was exposed to the 35 W lamp, photons were absorbed, leading to the formation of electron-hole pairs. Upon excitation, the h^+ and e^- later came into contact with O_2 and H_2O forming radicals. The formed radicals were able to destruct the chromophoric groups from the dye molecules, resulting in smaller, less toxic colored compounds. Upon testing for reusability, the material could be re-used with a 60% efficiency after five cycles. The loss of efficiency could be due to surface fouling, structural changes, and limited number of active sites.

Though several studies have been conducted using green IONPs, the degradation is generally above 70% even though there is no direct trend in terms of the degradation efficiency (Table 2). Several studies are looking at studying the traditional reaction parameters, such as pH, concentration, dosage, temperature, and time to obtain full efficiency. For

example, not a lot of studies reported on the effect of pH, which is one of the important surface characteristics. This surface characteristic of pH can facilitate effective adsorption of the anionic dye molecules, attributed to the strong Lewis acid-base interactions. In a study by Ahmed et al. [3], the effect of pH was investigated. The highest degradation efficiency of 94% against AYR dye at an optimal pH of 6 was noted, highlighting the critical role of pH in influencing surface charge and dye-catalyst interactions.

Understanding another variable such as concentration, it is known that the nature and concentration of the target dye plays a role in the photocatalysis process [58]. Organic dyes serve as photosensitizing chromophores in dye-sensitized photocatalysis. Upon absorbing visible light, these dye molecules become photoexcited and inject electrons from their excited state into the conduction band of a semiconductor possessing an appropriate band gap, effectively extending the light absorption range of the material [37]. This allows the IONPs as a semiconductor to experience electronic excitation even under visible light irradiation, generating electron-hole pairs through redox reactions. The injected electrons further contribute to charge carrier dynamics by replenishing conduction band electrons, facilitating sustained photocatalytic activity. The chromophore (azo, di-azo, phenothiazine, and many more) and the type of IONPs (hematite, maghemite and magnetite) as a semiconductor interaction thus enhances light harvesting and promotes efficient electron transfer, acting both as a light absorber and an intermediary for photocatalytic processes [40,59]. For an example, a basic dye like Azo = CV and phenothiazine = MB dyes will interact with IONPs surface by using π - π interactions and electrostatic attraction [43,44].

At low dye concentrations, photocatalytic degradation by IONPs tends to be more efficient due to minimal competition for active sites on the catalyst surface. Therefore, the solution remains relatively transparent, which facilitates deeper light penetration and allows more photons to reach the catalyst, enhancing its photoexcitation and promoting efficient generation of reactive oxygen species (ROS), such as hydroxyl ($\bullet$ OH) and superoxide ($\text{O}_2\bullet^-$) radicals [41]. This leads to rapid and effective dye degradation. Concurrently, the inner filter effect becomes more pronounced; excess dye molecules in solution absorb a significant portion of the incident light before it can reach the photocatalyst surface, diminishing photon-induced charge excitation and limiting overall photocatalytic activity [51]. As a result, even with a higher concentration of dye, the degradation rate may decline rather than improve.

The dosage of IONPs plays a critical role in determining photocatalytic efficiency during the degradation of various dyes [60]. An insufficient catalyst dosage may lead to reduced degradation performance due to the limited availability of active surface sites, whereas higher dosages generally enhance photocatalytic [37]. This enhancement occurs because an increased catalyst concentration provides more surface area for dye adsorption, such as in the case of methylene blue (MB), facilitating greater interaction between dye molecules and the nanoparticle surface. Additionally, a higher concentration of IONPs in the reaction medium improves light absorption, thereby increasing the number of nanoparticles that are photoactivated efficiency [56]. This results in the generation of more electron-hole (e^-/h^+) pairs and, consequently, a higher production of radicals, which drive the degradation process more effectively [57].

5. Limitations and Resolutions in the Photocatalytic Use of Green IONPS

Despite their environmental compatibility and affordability, IONPs exhibit several inherent limitations that hinder their standalone photocatalytic efficiency. Among the general drawbacks are particle agglomeration, low photostability, surface defects, and limited light absorption in the visible range [52]. These issues compromise active surface area and charge transport, leading to diminished degradation performance. When evaluating

specific iron oxide phases, further limitations are observed. Hematite ($\alpha\text{-Fe}_2\text{O}_3$) suffers from poor electrical conductivity and rapid recombination of photogenerated electron–hole pairs, which significantly lowers its photocatalytic efficiency, particularly under visible light [61,62].

From Table 3, it can be noted that modification with carbon materials for enhanced surface area, doping with low bandgap metals to reduce the recombination rate and forming various heterostructures through green materials was conducted to improve the surface and optical properties of these IONPs. Tumbelaka et al. [63] using the co precipitation method in the presence of *Moringa olifera* formed various ratios of non-uniform and dispersed $\text{Fe}_3\text{O}_4/\text{TiO}_2$. Upon testing their efficiency, for the ratio of 1:7, a 99.9% degradation of MB after 2 h was recorded. Furthermore, the materials were reused more than four times, producing excellent stability. Ramadhani et al. [64] upon loading different concentrations of sheets of rGO on Fe_3O_4 recorded a reduced bandgap of 2.7 eV and a small particle size of 9.3 nm. A high RhB dye degradation of 98% was noted. Fauzi et al. [65] studied the effect of dosage (0.02–0.1 g) against MB dye using the $\text{Fe}_3\text{O}_4\text{-TiO}_2$ composited formed via *M. olifera* extract. A material with a bandgap of 3.48 eV produced an efficiency of 98% against MB in 60 min, using an optimum dosage of 0.06 of catalyst. Above this loading, agglomeration of the catalyst occurred. It could also be that the small particle size of 5.4 nm could have assisted with the high degradation rate and these green composites could be re-used up to two times with high efficiency. Sari et al. [66] formed semi spherical agglomerated $\text{Fe}_3\text{O}_4/\text{C}$ -dots NPs via *Moringa olifera*. A decreased magnetism was noted, upon incorporating C-dots from 59 emu/g to 29.2. Moreover, an increased band-energy was also noted with the increase in concentration of the presence of carbon. Upon testing their catalytic efficiency, a 98% MB degradation in 10 min under UV light was recorded. The increased activity at @ 25 mL C-dot concentration was as a result of the increased active sites from the carbon materials, even though at higher concentrations (30 mL) and prolonged periods (30 min), saturation on the nanocomposite occurred. From trapping experiments with OH radicals and h^+ were noted to being the species most responsible for the degradation. Moreover, these materials could be re-used up to three times with a steady decrease in efficiency from 98 to 50%. This was attributed to mass loss of the catalyst after every cycle. From Figure 8a,b, EDS and mapping confirmed that the structure of the catalyst was still intact, with the main peaks of Fe, O, and C still in existence. This was further corroborated by bonding analysis via FTIR (Figure 8c), confirming the existence of the main functional groups after several reuses.

Palanivel et al. [67], formed a Z scheme ternary heterojunction using the hydrothermal method through green tea extract. The low bandgap of 2.83 eV assisted in the 97% MB degradation. The authors noted that ternary materials tend to produce greater degradation. Moreover, the Z scheme transfer helps with the charge carrier's recombination rate. Also, the rGo supported heterostructure increased the number of active sites through improved surface area ($304\text{ m}^2/\text{g}$). This was due to the functional groups present in the carbon material. Prescillio et al. [68] formed a low bandgap (1.08 eV) heterojunction of $\text{FeO}/\text{ZnO-Ni}$ from *Magonia pubescens* extract. A 97% RhB dye degradation was recorded. The authors, through the DFT study, noted that the p-n heterojunction formation assisted with the reduced recombination rate and the defects. Moreover, the doping with Ni^{+} species, lowered the bandgap, introducing defects caused by the d-orbital contributions. Rini et al. [69] formed a Z-scheme heterojuncture of *Amaranthus viridis* leaf extract of $\text{Fe}_3\text{O}_4/\text{rGO}/\text{ZnO}$ (Figure 9). After 180 min using a 30 W UV lamp, with a bandgap of 2.62 eV, a 90% degradation was observed at an increased ratio of 1:5. Also, a redshift was noted, increasing the number of defects and selective mobility. Lastly, the heterostructures could be reused up to three cycles without losing efficiency and the electrons and holes

were the species most responsible for the high degradation. The authors also stated that the e^- were captured through the rGO carbon sheet within the heterojunction (Figure 10). This prevented the recombination of the electrons and holes. These separated species ideally interacted with the $O\cdot$ and OH^- on the surface forming radicals. These formed radicals were therefore used to breakdown the dye chromophores and pollutants.

Table 3. Green IONPs composites for dye degradation.

Composite Material	Target Dye	Degradation Conditions	Optical Properties	Efficiency and Time	Reference
Ag-Fe ₂ O ₃ (<i>Saussurea obvallata</i>)	MB	10 mg, 10 ⁻⁵ M	3.26 eV, 380 nm	97%, 60 min	[52]
Ag-Fe ₂ O ₃ <i>Cabbage peel</i>	PR			78%, 8 min	[61]
Ag-Fe ₂ O ₃ <i>Kulekharal leaves</i>	CV MG	100 mg/L, 20 uL	429 nm	100%, 3 min	[53]
AgFe ₂ O ₃ , Ag-AC Fe ₂ O ₃ @Ag-@AC <i>Cocos nucifera</i> . L.	MB	0.25 and 0.5 g	3.73 eV 3.75 eV 3.69 eV	100%, 30 min	[54]
Fe ₂ O ₃ -Ag-ZnO <i>Poinciana Leaf</i>	RhB	1.0 g/L, 10 ppm, pH-10		99%, 45	[55]
Fe ₃ O ₄ /TiO ₂ <i>green via Moringa extract</i>	MB	-	~2.76 eV 197 nm	99.9%, 120 min 4 cycles	[63]
Fe ₃ O ₄ /rGO	RhB	0.05 g	2.7	98%	[64]
Fe ₃ O ₄ /TiO ₂ <i>Moringa extract</i>	MB	0.02–0.1 g	3.48 eV 384, 396 nm	98%, 60 min	[65]
Fe ₃ O ₄ /Cdots <i>Waterlemon peel and M. olifera</i>	MB	50 mg, 7 ppm, 30 W UV lamp	3.44 eV	10 min	[66]
Fe ₂ O ₃ -g-C ₃ N ₄ -rGO <i>Tea extract</i>	MB		2.83 eV	97%	[67]
FeZnO FeZnO-Ni <i>Magonia pubescens</i>	RhB	Visible light	1.08 eV	87%, 97%, 300 min	[68]
Fe ₃ O ₄ /rGO/ZnO <i>Amaranthus viridis leaf</i>	RhB	30 W UV, 50 mg, 7 mg/L	2.6 eV	90%, 180 min	[69]

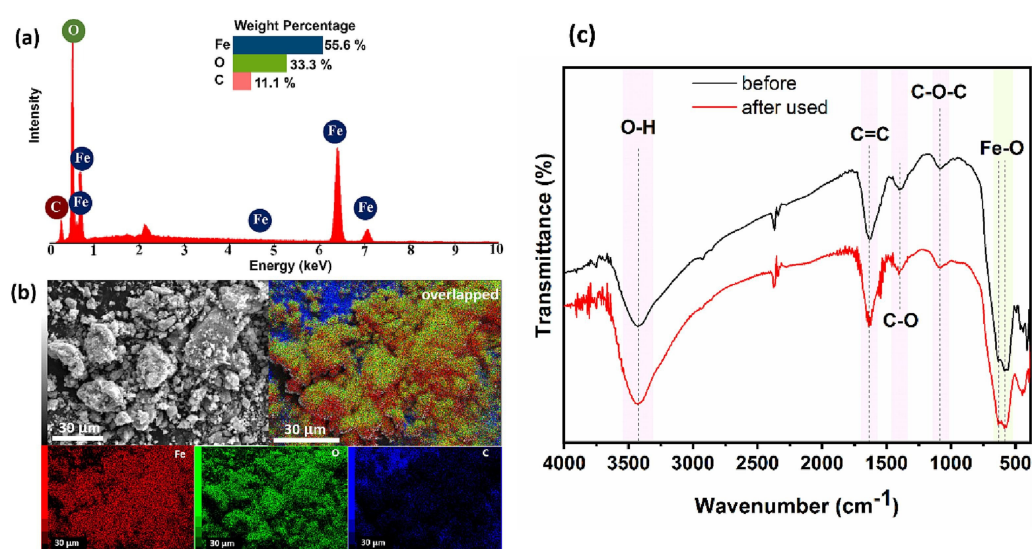


Figure 8. (a) EDS, (b) Mapping and (c) FTIR of MO-WM Fe₃O₄/C-dots (OA) Reprinted from [66].

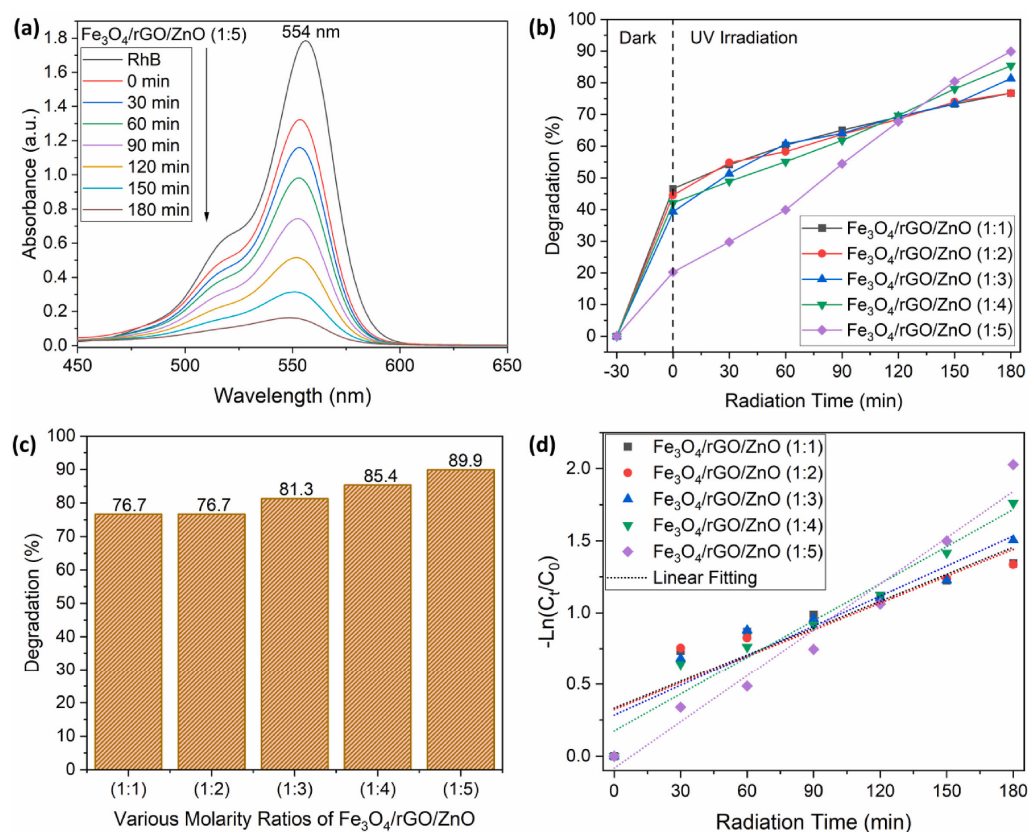


Figure 9. (a) Absorption spectra, (b,c) percentage degradation (%) and (d) kinetics of green Fe₃O₄/rGO/ZnO against RhB dye (OA) Reprinted from [69].

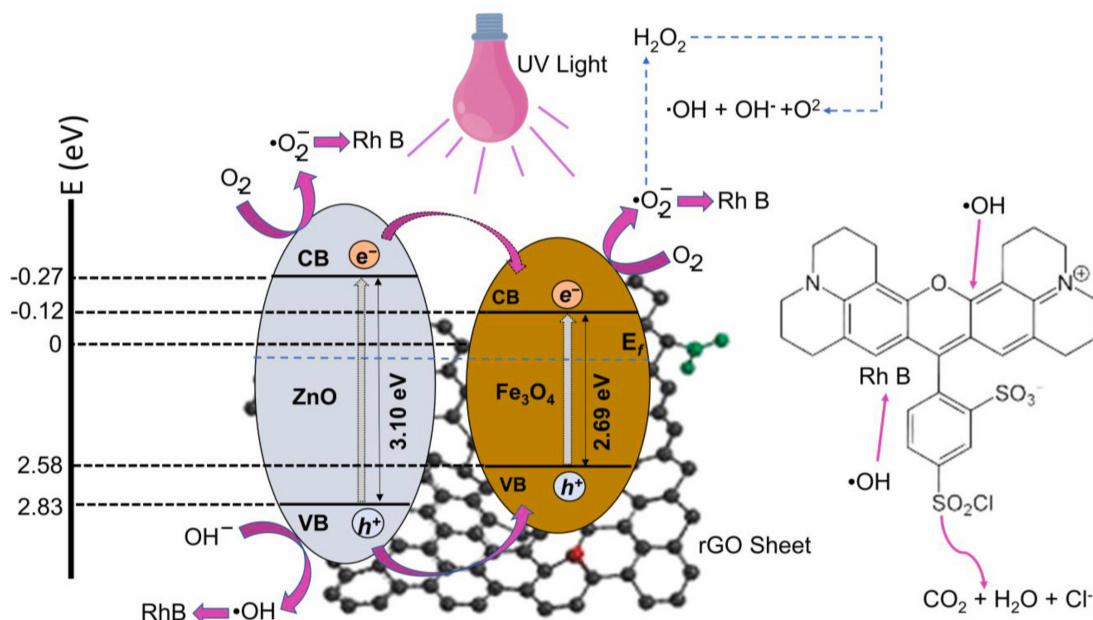


Figure 10. Mechanism of degradation against RhB dye in the presence of UV light using Fe₃O₄/rGO/ZnO (OA) Reprinted from [69].

Also, outside of forming heterostructures, authors have noted through synthesis, tailoring, and loading metals and non-metals such as Ag, Au, Pt and Cu, respectively, can enhance the degradation efficiency [58]. Ag is the most preferred and highly used doping metal used since it is cost effective. The deposition of the metals and non-metals facilitates the surface plasmon resonance, as these metals can form reactive species, give access to

the visible range, and improve the surface area. These enhancements are caused by the introduction of defects via surface modification and lattice distortion. As noted in Table 3, several studies introduced Ag on IONPs and in all these studies, enhanced degradation was noted. Matos et al. also showed from *Mytenus*-mediated Fe_2O_3 surface defect through plant phytochemicals could be induced (Figure 11). They formed a defect enhanced visible light photocatalysts, introducing oxygen and iron vacancies, which assisted with improving the charge separation and forming reactive oxygen species, thus a high degradation and reusability of the material was noted.

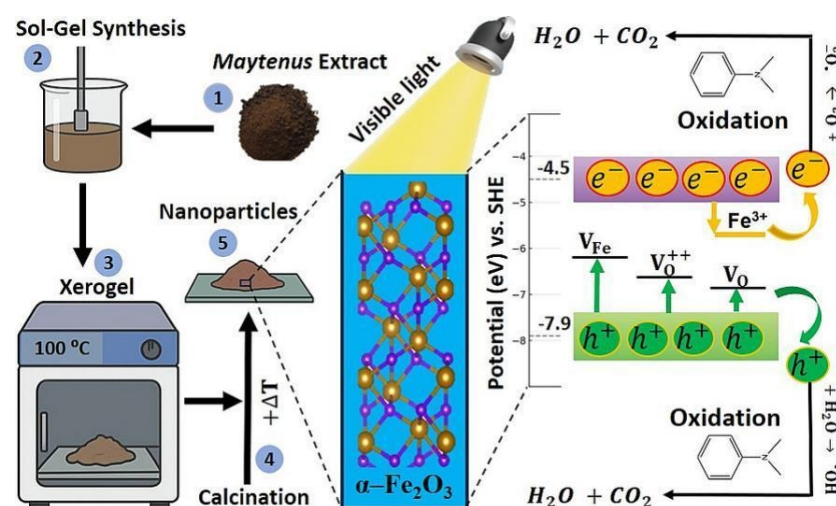


Figure 11. *Maytenus* mediated IONPs synthesis and application as photocatalysts [70].

6. Conclusions

Current treatment methods in the market are not able to remove dye pollutants completely. Secondary pollutants through leaching tend to form, which can be more toxic than primary pollutants. Also, their formation of sludge, inability to remove a spectrum of pollutants and constant need for maintenance has led to the usage of photocatalysis. Due to these limitations, it is important to find safer alternatives in an effort to protect the ecosystem, the environment, and human health. For the photocatalysis method to be efficient, a green and suitable IONP catalyst is required. The photocatalysis method has offered several advantages over conventional removal techniques. It has been shown to degrade both organic and inorganic pollutants. During this process, more than 80% of the targeted pollutants can be degraded. However, efficiency of this method, in particular for green materials, is still required for high modification. This includes studying the specific phytochemicals, pH, time, temperature to assist in the formation of the required morphological, and optical and surface properties of the materials formed. Moreover, as shown in Table 3, even upon doping or forming heterostructures, in certain instances high degradation efficiency still occurred at prolonged period (60–300 min).

Therefore, even though multiple reports on green IONPs have been reported, more research needs to be conducted to improve their performance. These include the following:

- i. Reusability and stability studies on green IONPs materials, as this will affect the cost viability of these materials, hence they need to be conducted;
- ii. EDS, SEM, XRD and FTIR can be used to analyze the structure of these catalysts for before and after degradation analysis;
- iii. More understanding on the deposition of low bandgap materials or formation of heterostructures. Though improvement on low bandgap reduction was shown and

- increased surface area leading to more active sites, several studies still reported on results similar to single metal oxide NP for degradation;
- iv. Doping with metals and non-metals can help with the tuning of the electronic structure of these materials to ensure there is access to the broader visible spectrum;
 - v. Through DFT and PL studies, understanding and introducing a controlled number of defects to the system can also assist with improved active sites;
 - vi. During synthesis, studying the effect of plant concentration for the controlled morphology, size, and porosity is important to ensure there is no over or under capping, as that might also lead to highly agglomerated materials.

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Abbreviations

The following abbreviations are used in this manuscript:

IONPS	Ion Oxide Nano Particles
OA	Open Access
GC-MS	Gas Chromatography coupled with Mass Spectrometer
A-3	<i>A. fumigatus</i>
L-6	<i>P. fluorescens</i>
RO	Reverse Osmosis
NF	Nano Filtration
ZnO	Zinc Oxide
TiO ₂	Titanium dioxide
NiO	Nickel Oxide
γ-Fe ₂ O ₃	hematite
α-Fe ₂ O ₃	Maghemite
Fe ₂ O ₃	Magnetite
FeO	Ferrous Oxide
FeNPs	Iron Nano Particles
FeCl ₂	Iron II Chloride/Ferrous Chloride
MB	Methylene Blue
CV	Crystal violet
AYR	Alizarin yellow R
RR	Remazol yellow
S	Safranin
RB	Reactive blue
RhB	Rhodamine Blue
FTIR	Fourier Transform Infrared Spectroscopy
XRD	X-ray diffraction
CR	Congo Red
MO	Methylene Orange

EB	Evans Blue
PR	Phenol Red
$O_2^{\cdot -}$	superoxide radicals
$OH^{\cdot}$	Hydroxyl radicals
EY	Eosin Yellow
$Fe(NO_3)_3 \cdot 9H_2O$	Iron(III) nitrate nonahydrate
$FeCl_3 \cdot 6H_2O$	Iron(III) chloride hexahydrate
$FeSO_4 \cdot 7H_2O$	Iron(II)
$NaBH_4$	Sodium borohydride
$N_2H_4 \cdot H_2O$	Hydrazine hydrate
NH_3	Ammonia
NH_4OH	Ammonium hydroxide
NaOH	Sodium hydroxide
EDA	Ethylenediamine

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